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Simulation of Nitrate Biogeochemistry and Reactive Transport in a California Groundwater Basin

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Nitrate is the number one drinking water contaminant in the United States. It is pervasive in surface and groundwater systems, and its principal anthropogenic sources have increased dramatically in the last 50 years. In California alone, one third of the public drinking-water wells has been lost since 1988 and nitrate contamination is the most common reason for abandonment. Effective nitrate management in groundwater is complicated by uncertainties related to multiple point and non-point sources, hydrogeologic complexity, geochemical reactivity, and quantification of denitrification processes. In this paper, we review an integrated experimental and simulation-based framework being developed to study the fate of nitrate in a 25 km-long groundwater subbasin south of San Jose, California, a historically agricultural area now undergoing rapid urbanization with increasing demands for groundwater. The modeling approach is driven by a need to integrate new and archival data that support the hypothesis that nitrate fate and transport at the basin scale is intricately related to hydrostratigraphic complexity, variability of flow paths and groundwater residence times, microbial activity, and multiple geochemical reaction mechanisms. This study synthesizes these disparate and multi-scale data into a three-dimensional and highly resolved reactive transport modeling framework.

1. BACKGROUND

Nitrate contamination is pervasive in surface and groundwater systems and is a growing problem in California. Greater than 40 percent of the State's population uses groundwater for at least a portion of their domestic needs, and some cities, such as Fresno, Davis, and

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Lodi in the Central Valley, rely solely on groundwater. Since 1984, roughly 8,600 out of 25,000 wells in the State have been shut down, primarily because of nitrate contamination. About 10 percent of the currently operating public wells produce water that exceeds the regulatory drinking water standard (10 mg/L as N), and a much larger fraction produce water approaching this standard. As the population increases over the next 20 years, the loss of groundwater resources due to nitrate contamination will become an increasingly severe water supply issue [1]. Nitrate issues also affect the planned use of groundwater basins to store water in lieu of above-ground reservoirs [2].

Nitrate contamination is commonly viewed as intractable because it is ubiquitous, has multiple sources, and is expensive to treat. Contamination of groundwater is particularly problematic because aquifers have long response times (years to decades), and are heterogeneous and difficult to characterize. In California, the main anthropogenic activities that contribute nitrate to groundwater – fertilizers, confined animal feeding operations, and septic systems – are a legacy of commerce and growth over the last half-century, yet remain vital to the economic future of the State. Notably, the actual economic impacts of contamination, including the need to blend or treat water supplies, abandon wells, or secure alternative sources, are not well documented or understood.

Management of nitrate contamination requires quantitative assessments of (1) the source, distribution, and evolution of nitrate concentrations in affected aquifers, (2) the economic dimensions of the problem to implement cost-effective remediation, and (3) impact of land and water management practices that have been designed to reduce nitrate loading in order to implement effective source mitigation. All three goals require a fundamental understanding of fixed nitrogen transport and chemistry in the saturated zone on a basin scale.

The biogeochemical cycling of nitrogen between source areas near the ground surface and the vadose and saturated zones is complex and can be microbially mediated [3–6]. In oxic groundwater regimes, nitrate is anionic with no appreciable sorption. Microbial denitrification in the saturated zone facilitates the conversion of nitrate to dissolved molecular nitrogen (N_2), and is the ultimate sink for nitrate under low oxygen conditions. Characterizing the microbial controls and the kinetics of denitrification is essential to developing accurate reactive transport models for nitrate in groundwater. Developing such models also requires the ability to accurately characterize and model groundwater flow paths in heterogeneous media at both the field scale and basin scale.

2. THE LLAGAS SUBBASIN

This paper reviews the initial development of a basin-wide groundwater flow and transport model to study migration and fate of nitrate in the Llagas groundwater subbasin, situated in a narrow inland valley approximately 20 km south of San Jose, California (Fig. 1). Nitrate contamination of shallow groundwater is pervasive in many parts of the subbasin as a result of numerous rural and agricultural land uses [8,9]. Over the past

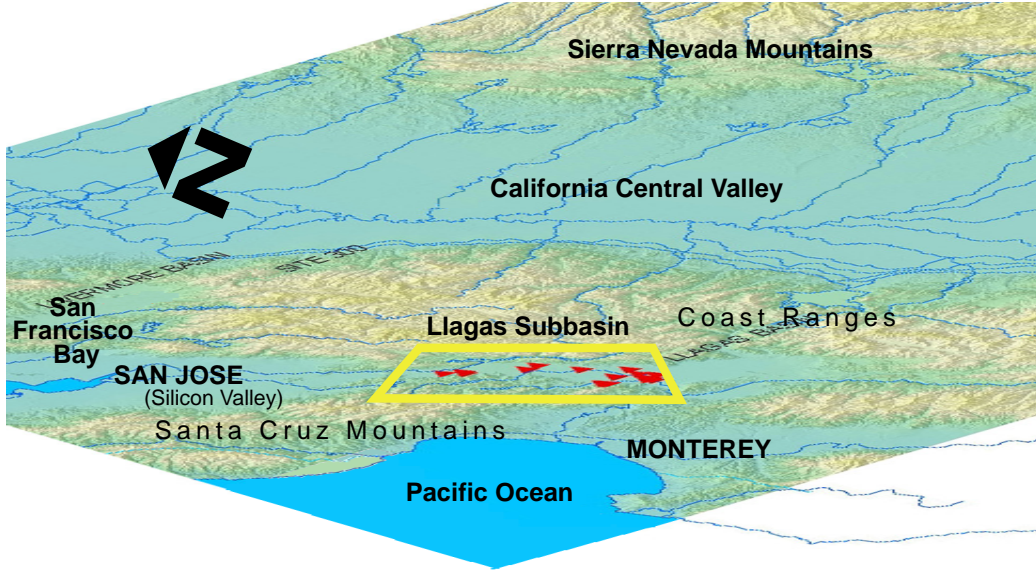


Figure 1. Bird's eye view of the Llagas subbasin location south of San Jose, California, looking northeastward.

50 years, these valley areas have undergone significant growth and urbanization due to their proximity to Silicon Valley, and groundwater provides the only source of water to some areas in the subbasin. The shallow zones are also threatened by widespread perchlorate contamination. Contamination in the shallow zones threatens deeper zones where increased water production is expected in the future to meet the demands of an increasing local population.

At its northern end, the subbasin extends from a groundwater divide (at its junction with the Coyote subbasin) to the south where it is bounded by the Pajaro River [7]. The subbasin is approximately 25 km long and ranges between 5 and 8 km wide. Annual precipitation ranges from less than 40 cm in the south to more than 60 cm in the north. Water level elevations (Fig. 2) indicate a southeasterly groundwater flow direction with significant natural recharge occurring where streams discharge into the subbasin south and northeast of Morgan Hill and west of Gilroy [10].

3. MODELING STRATEGY AND FRAMEWORK

Our reactive transport modeling approach focuses on the development of a hierarchical simulation framework that can be used as a means to assess the impacts of spatially variable nitrate loading, nitrate transport, and reactive (denitrification) processes between the water table and both shallow and deeper groundwater zones. Our simulation strategy

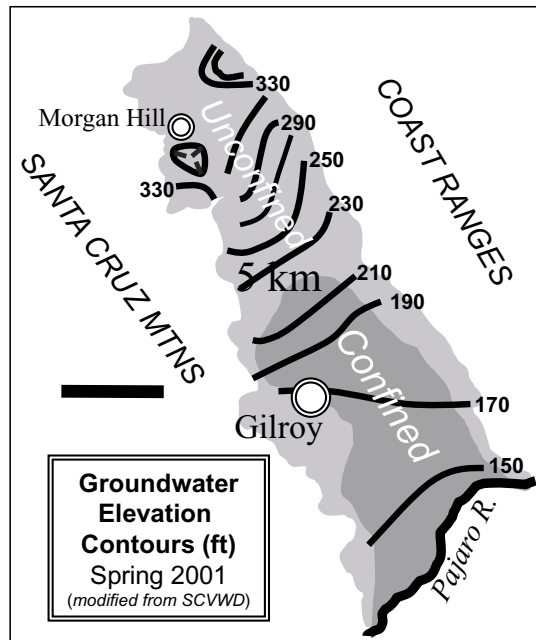


Figure 2. Map of Llagas subbasin showing groundwater elevation contours (in feet above sea level) for Spring, 2001. Modified from [10].

will be iterative and grounded, initially, in the development of a detailed geologic model. As discussed further below, flow model development will be guided by isotopic age dating measurements, noble gas tracer measurements, inverse geochemical models, and additional biogeochemical characterization of denitrification processes, as determined from both the field and laboratory. Ultimately, model simulations will allow the evaluation of important sensitivities and factors that control the flux, distribution, and residence times of nitrate in the subbasin and, thus, may form a basis for improving basin nitrate management strategies in the future.

3.1. Geostatistical Simulation of Hydrostratigraphy

The water bearing formations of the Llagas subbasin include Pliocene to Holocene age deposits of unconsolidated to semi-consolidated gravel, sand, silt and clay [7]. These include the deeper Santa Clara Formation and alluvial and alluvial fan deposits, which constitute the principal water producing zones. This and previous hydrogeologic interpretations [7,11] depict the hydrostratigraphic architecture as consisting of thick, gently dipping, and laterally continuous layers composed of three principle hydrofacies: (1) coarse-grained materials, (2) fine-grained materials including lacustrine deposits, and (3) alternating thin and discontinuous layers gravel and silt or clay. In a hydrogeologic context, these three hydrofacies can be regarded as aquifers, aquitards, and aquicludes, respectively.

Geostatistical simulation methods can be used to generate representative three-dimensional “realizations” of alluvial hydrofacies architecture [12–14]. In this study, realizations are designed to replicate the patterns of heterogeneity evident in the Llagas subbasin aquifer system. These patterns are deduced from geologic cross sections [7,11] and direct measurement of vertical transition probabilities from high-quality lithologic logs. Spatial variability of hydrofacies is quantified through a transition probability Markov approach [12–14]. The realizations are conditioned by lithologic logs, including driller’s logs, and can be adjusted to reflect larger scale structural patterns or trends such as dips or transitions between formations, e.g., [16].

The richest source of data that describe hydrofacies architecture is found in drillers’ logs, which, understandably, can be of variable quality. Over 300 drillers logs (Fig. 3) have been used to condition the geostatistical realizations developed in this study (Fig. 4). The size of the domain in this representation is 24 km (x) \times 8 km (y) \times 0.3 km (z). The spatial resolution is 100 m in the horizontal directions (x, y) and 2 m in the vertical (z), yielding over 2.8 million blocks in the model realization. Conditioning of the geostatistical realizations to drillers’ logs data is accomplished through a novel technique that accounts for data inaccuracy through assigned correlations between the data and true lithology [15]. For example, if a particular driller’s log is assumed absolutely correct in describing lithology, the assigned correlation is 1.0. Likewise, if the driller’s logs is assumed completely inaccurate, or “random”, the assigned correlation is 0.0.

The geostatistical simulation algorithm attempts to preserve spatial correlation inferred

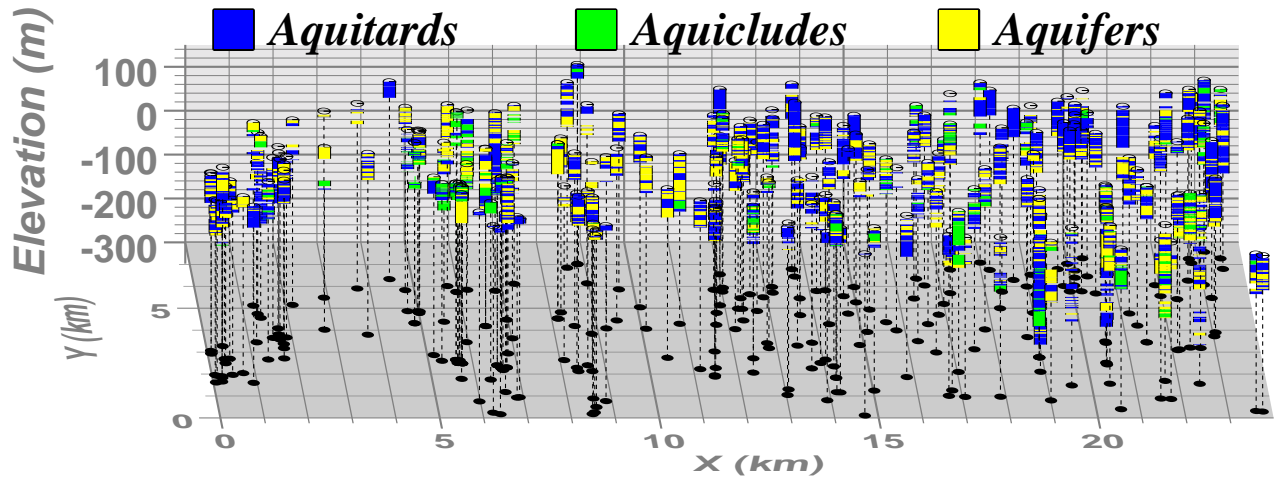


Figure 3. Driller's log lithologic data categorized as aquifers, aquitards, and aquicludes in Llagas subbasin (looking northeastward).

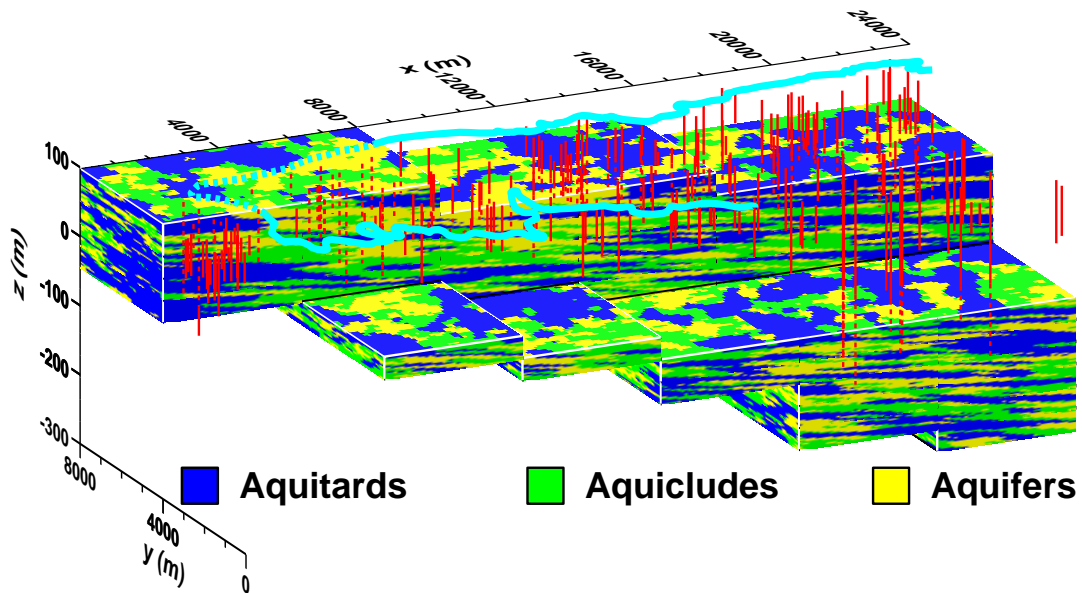


Figure 4. Example geostatistical realization of hydrofacies architecture in the Llagas subbasin, looking northeastward. The domain is 24 km long, 8 km wide, and 0.3 km deep, with a spatial resolution of 100 m in the horizontal directions and 1 m in the vertical. Red lines represent wells in Fig. 3. Light blue outline represents the subbasin boundary at the surface.

from the hydrofacies data; thus drillers' log data are honored to varying degrees depending on data quality. In this manner, the better quality data exert a stronger influence on the realizations. Conversely, poor quality data do not force unrealistic stratigraphic relationships. In application to the Llagas subbasin lithologic data, assigned correlation for different driller's logs ranges between 0.3 to 0.7, depending on quality of lithologic description. Assigned correlation of 1.0 was ascribed only to lithologic logs carefully described by hydrogeologists.

3.2. Isotopic Age Dating and Source Identification of Groundwater

Groundwater "age" represents the mean amount of time a groundwater parcel has resided in the saturated zone, isolated from any connection with the atmosphere. Groundwater ages can be used to understand travel times along flow pathways between recharge locations and sampling points, which are typically wells [16]. The water parcels collected in such a sample may be widely distributed in terms of their ages, largely as a function of the sampling length of the screened interval and dispersion and diffusion along flow pathways that reach the well [16].

Groundwater ages can be estimated from a combination of isotopic dating approaches: (1) tritium- ^3He ratios, based upon the amount of ^3He ingrown from the radioactive decay of tritium (an isotope of hydrogen incorporated in some water molecules), can be used to estimate the age of groundwater less than 50 years old; (2) raw tritium concentrations can be decay-corrected according to the tritium-helium age and compared with historical tritium concentrations in precipitation to indicate the fraction of water over 50 years old, and (3) radiogenic ^4He concentrations derived from the radioactive decay of uranium and thorium minerals in the subsurface can be used to estimate the age of the fraction of water over 50 years old.

In the Llagas subbasin, tritium- ^3He and ^4He groundwater age estimates were obtained from 13 wells (Fig. 5). In the Morgan Hill area, older tritium-helium ages generally correspond to older ^4He ages (Fig. 6). The younger tritium- ^3He age dates further refine wells vulnerable to water quality impairment. A sample from southeast Gilroy containing the youngest groundwater analyzed thus far may be influenced by recharge immediately to the west. Age versus depth relationships are difficult to infer because of long and multiple well screen intervals but stratified groundwater (younger, contaminated groundwater in the shallow section) is evident in wells with differing perforation intervals.

Several techniques can be used to infer the source of groundwater at a sampling point, or the source of nitrogen within the groundwater. For example, artificial recharge is being implemented in the northern portion of the Llagas subbasin using imported water from the San Francisco Bay Delta, which is fed by Sierra Nevada river flows. Because this water is depleted in its content of the ^{18}O isotope (incorporated in some water molecules), it is isotopically lighter than the recharge derived from local precipitation, and can be readily distinguished in groundwater. Recent ^{18}O measurements indicate that imported water is present in 6 of 12 wells sampled in the Morgan Hill area, while other wells in the Llagas

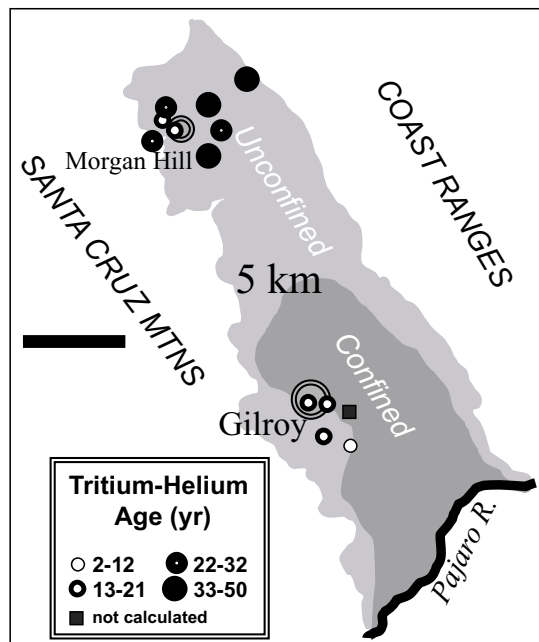


Figure 5. Tritium-helium groundwater age estimates for 13 samples obtained from Llagas subbasin.

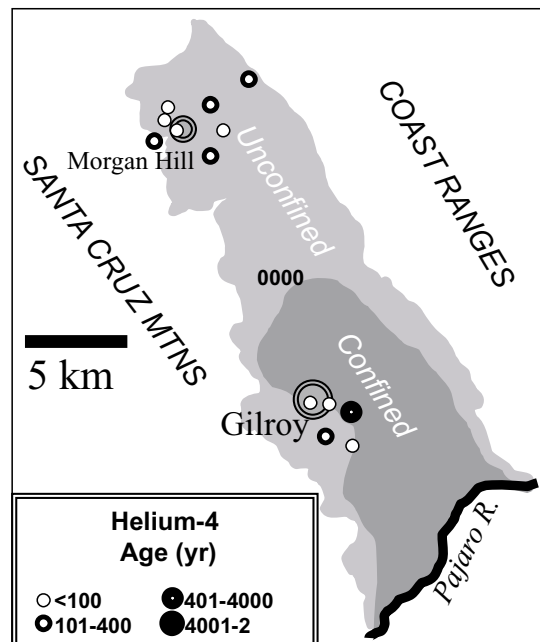


Figure 6. Helium-4 groundwater age estimates for 13 samples obtained from Llagas sub-basin.

subbasin indicate exclusively local recharge sources.

Measurements of dissolved air in groundwater samples (chiefly, Ar, N₂, and O₂) and the fraction of the nitrogen isotope ¹⁵N present can also be used to assess whether excess nitrogen exists as a product of microbial denitrification along an upgradient flow path, as well as correlate nitrate nitrogen with potential nitrate sources above the water table.

Altogether, groundwater age estimates larger or smaller than 100 years can help distinguish groundwater sources subject to anthropogenic effects. The use of groundwater age dates along with estimates of recharge sources, dissolved air, and dissolved nitrogen can help identify realistic lateral and vertical flow paths and travel times for flow models, as well as document the existence of denitrification and potential sources of nitrate along these flow paths. The flow models, in turn, may provide insights on actual age distributions and depth relationships to improve interpretation of isotopic age estimates [16]. This synergy is useful for identifying aquifer zones susceptible to contamination, including the past and future impacts from nitrate loading.

3.3. Geochemical Inverse Modeling

Geochemical inverse modeling can be useful in relating groundwater quality observations at specific locations to potential reactions occurring along upgradient flow paths or streamlines. For example, the inverse component of the PHREEQC geochemical modeling code [17] uses mass balance constraints imposed on chemical reactions to infer the probability of an assumed reaction mechanism. In the Llagas subbasin, such geochemical constraints can be used to identify and check plausibility of reaction mechanisms along putative flow paths subject to nitrate loading. For example, oxidation reactions entailing suspected nitrate sources, such as animal fertilizers, will produce measurable changes in aquifer chemistry (e.g., increases in dissolved inorganic carbon, declines in pH, and elevated calcium and magnesium concentrations associated with carbonate mineral dissolution).

In Figure 7, nitrate concentrations show correlation with total calcium and magnesium concentration from several samples collected in the Llagas subbasin. The nitrate data appear to be clustered with respect to sodium concentration, a good indicator of total dissolved solids. The separation of the nitrate data with respect to total dissolved solids provides some means, albeit crude, for distinguishing different recharge sources and flow paths and, therefore, different nitrate sources. In the figure, three reaction mechanism models are compared that involve (1) organic fertilizer (e.g., C₅H₇O₂N), (2) chemical fertilizer (e.g., NH₄N₃), and (3) equal contributions of nitrogen from organic and chemical fertilizers on a molar basis (Combo). Comparison of the data to the models indicates that mixing of recharge water and nitrate loading subject to different reaction mechanisms contributes to the observed concentrations.

3.4. Microbial Processes

Microbially facilitated denitrification reactions will be incorporated into our transport model as field and laboratory data are gathered and interpreted. In addition to field

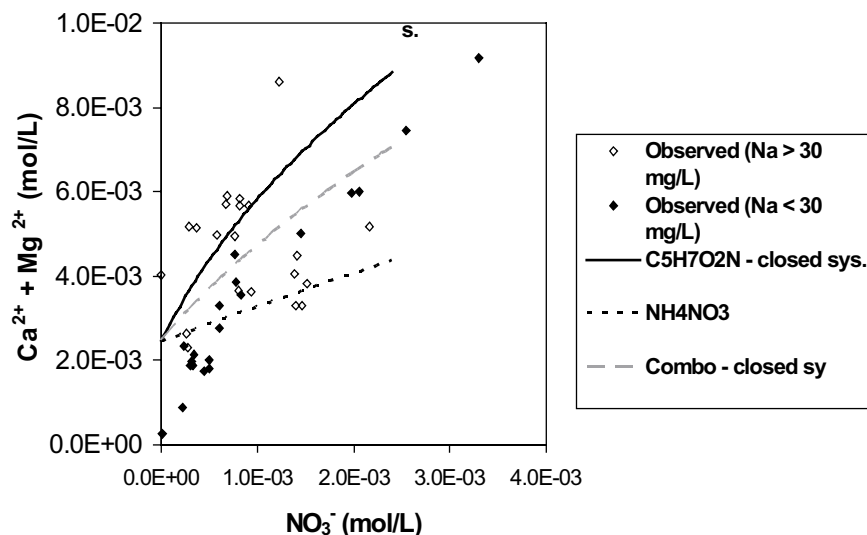


Figure 7. Comparison of nitrate to total calcium and magnesium concentrations, with possible nitrate source mechanism models.

data related to the identification of excess N_2 nitrogen in groundwater, two primary kinds of biological data will be used to address denitrification kinetics: (1) populations of denitrifying bacteria in aquifer samples collected in the field and (2) specific denitrification rates (i.e., mass of nitrate reduced per bacterial cell per unit time) determined in laboratory studies. To determine populations of denitrifying bacteria in field-collected aquifer samples, we will use quantitative, real-time PCR (Polymerase Chain Reaction) analysis that targets a diagnostic denitrification gene; based on an average of one gene copy per bacterial cell, the population of denitrifying bacteria in the sample can be calculated. Specific denitrification rates will be determined for the two major classes of denitrifying bacteria: heterotrophs and chemolithoautotrophs. Heterotrophic denitrifying bacteria use dissolved organic compounds as electron donors and as sources of cell carbon. Chemolithoautotrophic denitrifying bacteria, which can dominate denitrification activity in subsurface environments that are oligotrophic (i.e., low in organic carbon), use reduced inorganic compounds as electron donors and carbon dioxide as a source of cell carbon. As has been documented at numerous field sites, chemolithoautotrophic bacteria in aquifers often use reduced iron- and/or sulfur-containing minerals (such as FeS or FeS_2) as electron donors (e.g., [18]); such use of insoluble electron donors can render denitrification rates much slower for chemolithoautotrophs than for heterotrophs.

4. FLOW AND REACTIVE TRANSPORT MODELS

4.1. Hierarchical Flow Modeling Approach

Within the Llagas subbasin, two types of flow models will be considered. Initially, a subbasin-wide model will be developed in order to reconcile the basic water balances in the system (natural and artificial recharge, pumping and natural discharge, groundwater age, and water levels), both in terms of their current and historical configurations. This model will encompass confined and unconfined zones in the subbasin (e.g., as in Fig. 4), and will incorporate a vadose zone component to improve coupling of recharge processes to groundwater flow. The model will also be sufficiently detailed to resolve and distinguish the geostatistical variability of hydrofacies identified in Fig. 4 (2.8 million nodes). Codes such as Parflow [16,20] or NUFT [19] are being considered for this model.

More detailed, highly resolved submodels may be constructed in smaller portions of the subbasin volume, such as in the southern confined zone, as a means to more carefully study the effects of small-scale heterogeneity on transport and potential denitrification behavior. These models will be bounded by hydraulic conditions inferred from the subbasin model and will involve more highly resolved geostatistical detail within the three primary hydrofacies. Multi-million node simulations with Parflow will be required for these flow solutions.

Development and application of these models will be guided by isotopic age dating measurements, related groundwater and nitrogen source information, and inverse geochemical models. Apparent flow pathways inferred from these data must be reconciled with model simulations and the lithologic framework developed from the well observations. Conversely, the flow model and related transport simulations (below) will provide an important flow-path oriented framework for interpreting the isotopic measurements themselves, which are often derived from “mixed” water samples collected from wells with large screened intervals [16].

4.2. Hierarchical Transport Modeling Approach

Two types of transport models will be used in conjunction with both of the flow models mentioned in the previous section. On one level, particle based transport models (e.g., [21]) can be used to explore advection, dispersion, and time-of-flight (residence time) behavior for a small number of unreactive components in groundwater – such as nitrate or labeled recharge – and they can also be tailored to treat some types of simplified reactions, including denitrification.

To address more complicated behavior involving the transport and reaction of multiple dissolved species, especially in highly resolved flow systems, we will utilize a streamline-based reactive transport model. In this approach, a three-dimensional transport problem is recast into a large number of independent one-dimensional reactive transport simulations that correspond in a one-to-one fashion to a large number of streamlines that have been extracted from a three-dimensional flow field (e.g., [22,23]). The streamline mapping procedure involves a regridding process tailored specifically for transport simulations.

In many cases, this approach can drastically improve computational efficiency because (1) one dimensional transport problems are inherently easier to solve, (2) solutions on different streamlines can be obtained in parallel, and (3) computations may be focused in subsections of the flow domain where transport is of specific interest. In addition, the particular one-dimensional reactive transport model can be selected independently, such as PHREEQC [17], and used to represent, for example, aqueous complexation, oxidation-reduction processes, interphase transfer reactions (mineral precipitation and dissolution, ion exchange, and evolution of generated gases), and denitrification. The approach is largely limited to steady flow applications in which the importance of cross-streamline mass transfer (e.g., via transverse dispersion) is minimal.

4.3. Nitrate Migration and Fate

Ultimately, these models will be applied and tested iteratively to resolve and forecast nitrate loading and migration from one or more potential source areas, as documented in [8]. We expect to obtain improved perspectives and understanding of (1) the overall residence times of nitrate in the subbasin, especially as they pertain to selected source types, source locations, and proposed mitigation measures, (2) the potential role of denitrification as a sink mechanism for nitrate in the subbasin, in addition to natural discharge pathways, and (3) the potential for deeper zones of the subbasin to become contaminated with nitrate as a function of increased groundwater production in the future.

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